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(54) Abstract Title

Aggregates from fly ash

(57) Fly ash is mixed with a melting point lowering agent, a caking agent comprising bentonite, and with either a foaming agent comprising iron oxide, silicon carbide and carbonaceous material, or a material for adjusting the degree of reduction comprising carbonaceous material. The mixture is crushed so that the average particle size is up to 15 microns which is pelletized after which the pellets are baked within a temperature range of 1000°C ~ 1250°C.

The melting point lowering agent is made by mixing an alkali metal compound with Fly ash so that the total amount of Na<sub>2</sub>O and K<sub>2</sub>O respectively or both is within the range of 30 to 50 wt% in the mixture, melting the mixture at 1000° to 1200°C to form a glassy material and cooling and crushing the glassy material.

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Title of the Invention

Artificial Lightweight Aggregate and Manufacturing Method. Therefore

Field of the Invention

The present invention relates to a method of effectively using fly ash produced from coal fired boilers of coal fired power plants or the like, by recycling as an artificial lightweight aggregate for buildings, and for engineering public works, and to an artificial lightweight aggregate made thereby.

Background of the Invention

The effective use of fly ash produced from coal fired boilers of coal fired power plants or the like is a big problem.

As an effective use of fly ash, the use as an artificial lightweight aggregate which is in great demand, is suitable from the point of bulk disposal.

However, the use of fly ash produced from coal fired boilers of coal fired power plants as an aggregate, where part is aggregated by a sinter grate method, is still very limited.

The reason for this is that with coal fired boilers of coal fired power plants or the like, in order to reduce adhesion of the ash to the boiler pipes or the boiler wall, a coal which produces a high melting point ash is selected and used. That is to say, the fly ash generated from coal fired boilers of coal fired power plants or the like is

in general of a high melting point, and in order to make this into a lightweight aggregate this must be mixed with a large amount of low melting point clay or shale and baked. Securing this clay and shale in large amounts is difficult, and the mining, transportation, pre-processing, and mixing of this clay and shale, require a large expenditure, so that the manufacturing costs for the artificial lightweight aggregate are increased. Furthermore, since the utilization ratio of fly ash per unit product is low, then from the point of effective utilization of fly ash such use is not desirable. Moreover, the absolute dry specific gravity of the artificial lightweight aggregate using fly ash is approximately 1.2 ~ 1.4, and techniques for manufacturing a light artificial lightweight aggregate of an absolute dry specific gravity of 1.0 ~ 0.5 do not exist. Hence use is limited.

### Summary of the Invention

An object of the present invention is to provide a technique for producing at low cost a high specific strength high quality artificial lightweight aggregate at a comparatively low temperature, by adding a low cost additive which is easy to procure.

Furthermore, it is an object to increase the utilization ratio of fly ash per unit product and thus increase the utilization efficiency of fly ash, by reducing the amount of additive used.

Moreover, it is an object to increase the lightness and thus provide an extremely light artificial lightweight aggregate, to thereby extend its use.

### Description of the Preferred Embodiments

The artificial lightweight aggregate of the present invention is manufactured by mixing fly ash produced from a coal fired boiler with a melting point lowering agent, a caking agent, and a foaming agent to obtain a mixture, crushing the mixture so that the average particle size is up to 15 microns to obtain a pulverized product, adding water to the pulverized product and pelletizing to obtain pellets, and then baking the pellets in a rotary kiln within a temperature range of  $1000^{\circ}\text{C} \sim 1250^{\circ}\text{C}$  to give an absolute dry specific gravity of  $1.0 \sim 0.5$ . Furthermore, the method of manufacturing artificial lightweight aggregate according to the present invention involves; mixing fly ash produced from a coal fired boiler with a melting point lowering agent, a caking agent, and a foaming agent or a material for adjusting the degree of reduction, to obtain a mixture, crushing the mixture so that the average particle size is up to 15 microns to obtain a pulverized product, adding water to the pulverized product, pelletizing to obtain pellets, and then baking the pellets in a rotary kiln within a temperature range of  $1000^{\circ}\text{C} \sim 1250^{\circ}\text{C}$ . Here, drying may be carried out as required prior to baking.

The foaming agent comprises iron oxide in an amount such that the amount of  $\text{Fe}_2\text{O}_3$  in the fly ash is within the range of 1 wt. %  $\sim$  10 wt. %, carbonaceous material in an amount within the range of 0.2 wt. %  $\sim$  10 wt. % of the fly ash, and silicon carbide in an amount within the range of 0 wt. %  $\sim$  1 wt. % of the fly ash. In particular, in the case of baking to give an absolute dry specific gravity of  $1.0 \sim 0.5$ ,

then preferably iron oxide in an amount such that the amount of  $\text{Fe}_2\text{O}_3$  in the fly ash is within the range of 3 wt. % ~ 10 wt. %, silicon carbide in an amount of 0.1 wt. % ~ 1 wt. % of the fly ash, and carbonaceous material in an amount of 0.2 ~ 10 wt. % of the fly ash is added. The carbonaceous material is typically coal or coke.

The melting point lowering agent is made by mixing an alkali metal compound with fly ash so that the total amount of  $\text{Na}_2\text{O}$  and  $\text{K}_2\text{O}$  respectively or both is within the range of 30 wt. % ~ 50 wt. %, heating and melting within a temperature range of  $1000^\circ\text{C}$  ~  $1200^\circ\text{C}$  to form a glass, and then cooling and crushing.

The melting point lowering agent is then preferably added to the fly ash so that the total weight of the  $\text{Na}_2\text{O}$  and the  $\text{K}_2\text{O}$  is within the range of 2 wt. % ~ 6 wt. % in the baked product. Here the alkali metal compound is preferably sodium carbonate or potassium carbonate.

The reduction degree adjustment material is for adjusting the degree of reduction inside the aggregate, and is preferably made from carbonaceous material in an amount within the range of 0.2 wt. % ~ 10 wt. % of the fly ash. The carbonaceous material is typically coal or coke.

The present inventors added to fly ash as a melting point lowering agent, a product made by mixing an alkali metal compound with fly ash so that the total amount of  $\text{Na}_2\text{O}$  and  $\text{K}_2\text{O}$  respectively or both was 30 wt. % ~ 50 wt. %, heating and melting at  $1000^\circ\text{C}$  ~  $1200^\circ\text{C}$  to form a glass, and then cooling and crushing, so that the total amount of the converted amount of  $\text{Na}_2\text{O}$  and  $\text{K}_2\text{O}$  was 2 wt. % ~ 6 wt. %

of the baked product. As a result the melting point of the fly ash was lowered to a temperature of  $1000^{\circ}\text{C} \sim 1200^{\circ}\text{C}$  at which industrial wise, baking is relatively easy. Then, by adding iron oxide, silicon carbide, and carbonaceous material such as coal or coke with an average particle size of 10 microns or less as a foaming agent, a first artificial lightweight aggregate with a high specific strength and low water absorption at an absolute dry specific gravity of about  $0.5 \sim 1.5$  was baked.

Alternatively, by adding carbonaceous material such as coal or coke with an average particle size up to 10 microns as a reduction degree adjustment material in an amount of  $0.2 \sim 10$  wt. % of the fly ash, a second artificial lightweight aggregate with a high specific strength and low water absorption at an absolute dry specific gravity of about  $1.5 \sim 2.0$  was baked.

The iron oxide was added to the fly ash so that the amount of  $\text{Fe}_2\text{O}_3$  in the fly ash was  $1$  wt. %  $\sim 10$  wt. %. In particular, in the case where the absolute dry specific gravity was  $0.5 \sim 1.0$ , the  $\text{Fe}_2\text{O}_3$  was made at least  $3$  wt. % and silicon carbide was added to the fly ash to give  $0.1$  wt. %  $\sim 1$  wt. %. Furthermore, carbonaceous material in an amount of  $0.2$  wt. %  $\sim 10$  wt. % of the fly ash was used. Here the carbonaceous material also functions to adjust the reduction condition within the granulated pellet at the time of baking.

With a specific working example, the melting point lowering agent is produced by mixing sodium carbonate, potassium carbonate and carbonaceous material and then heating and melting at  $1000^{\circ}\text{C} \sim 1200^{\circ}\text{C}$  to form a glass where the total weight of the  $\text{Na}_2\text{O}$  and the  $\text{K}_2\text{O}$  respectively or both was  $30$  wt. %  $\sim 50$  wt. %

, and then cooling and crushing.

According to a method of manufacturing artificial lightweight aggregate of the present invention, at first, with respect to 100 parts by weight of fly ash being the raw material, bentonite being the caking material, is added to give 0.2 ~ 5 parts by weight in an external proportion, and the beforementioned melting point lowering agent is added so that the total of the converted amount of  $\text{Na}_2\text{O}$  and  $\text{K}_2\text{O}$  is 2 wt. % ~ 6 wt. % in the baked product.

Furthermore, in the case where the first artificial lightweight aggregate is obtained, the foaming agent is added in the beforementioned proportion. Alternatively, in the case where the second artificial lightweight aggregate is obtained, 0.2 wt. % ~ 10 wt. % of carbonaceous material such as coal or coke is added as the material for adjusting the degree of reduction inside the aggregate.

The mixture obtained in this way is then crushed to give an average particle size of up to 15 microns. The pellets are then obtained by pelletizing the pulverized product with water added. Subsequently, and after drying as necessary, the pellets are baked at  $1000^\circ\text{C}$  ~  $1250^\circ\text{C}$ .

The method of pelletizing used in the present invention, may be one which enables to give the pellets a predetermined diameter, being simply achieved using a pan pelletizer or an extrusion pelletizing machine. Moreover, for the baking, if continuous operation and uniformity of quality are considered, then use of a rotary kiln is preferable.

The melting point lowering agent is discussed hereunder.

With fly ash, it is commonly the case where the temperature at which a liquid phase is produced to initiate sintering is extremely high at  $1400^{\circ}\text{C} \sim 1500^{\circ}\text{C}$ . Baking the artificial lightweight aggregate at  $1400^{\circ}\text{C} \sim 1500^{\circ}\text{C}$  is not practical due to difficulties with fire resistance of the baking equipment, energy costs, and the selection of the foaming agent. Heretofore, in the case of baking such a raw material with a high degree of fire resistance, in general there is a method where... natural minerals such as clay or shale with a low degree of fire resistance and including a large amount of alkaline metal, or waste glass such as bottle glass etc. are added in large amounts as a melting point lowering agent. The various investigated results of the present inventors of the effect of adding clays, and shales, verified that in the constituents forming these clays and shales, the liquid phase forming temperature was remarkably low with alkali metals in a small amounts.

However, if an industrial chemical having a high included amount of alkali metals was added to the fly ash, only the surface of the pellet of granulated fly ash was melted, and the interior could not be baked. This is because the sodium and potassium salts being the industrial products of the alkali metals which are effective in lowering the melting point are mostly those which are water soluble, and in the step of drying the granulated pellet, the alkali metals are concentrated on the pellet surface so that at the time of baking, only the pellet surface is melted and the interior cannot be baked.

From the results of an investigation into a method of preventing concentration of the industrial alkali metals compounds on the pellet surface, it was found that

when a compound of an alkali metal such as sodium carbonate or potassium carbonate, was mixed with fly ash and heated and melted at  $1000^{\circ}\text{C} \sim 1200^{\circ}\text{C}$  to form a glass state so that the total amount of  $\text{Na}_2\text{O}$  and  $\text{K}_2\text{O}$  respectively or both was 30 wt. %  $\sim$  50 wt. %, and then cooled and crushed, and added to the fly ash so that the total amount of  $\text{Na}_2\text{O}$ ,  $\text{K}_2\text{O}$  was 2 wt. %  $\sim$  6 wt. % of the baked product, and then baked at  $1000^{\circ}\text{C} \sim 1250^{\circ}\text{C}$ , a high strength artificial lightweight aggregate foamed uniformly from the center was obtained.

With the present invention, the alkali metal compound such as sodium carbonate, or potassium carbonate used in the melting point lowering agent, is mass produced at low cost as an industrial chemical, and hence the present invention is advantageous cost wise. Furthermore, when the carbonate or the hydrogen carbonate of the alkali metal group is heated, harmful gas is not produced, and hence this is desirable. By adding this alkali metal compound to the fly ash, a glass which is difficult to dissolve in water is produced from the alkali metal group and silica. Since the fly ash is provided as silica source for producing the glass, then the fly ash can also be used in the melting point lowering agent. Hence the disposal rate of the fly ash can be improved, and new resources are not necessary, and hence this is desirable.

With the melting point lowering agent, with the total amount of  $\text{Na}_2\text{O}$  or  $\text{K}_2\text{O}$  respectively or both 30 wt. % or less, the melting point temperature for glassification exceeds  $1200^{\circ}\text{C}$  and hence the equipment and maintenance cost becomes high, and energy cost is also high. Furthermore, since the included percentage of alkali also

drops, the amount of melting point lowering agent used becomes great, and hence this is undesirable. Moreover, if the total amount of  $\text{Na}_2\text{O}$  or  $\text{K}_2\text{O}$  respectively or both exceeds 50 wt. %, then the water solubility of the formed glass increases so that only the surface of the granulated pellet is easily softened, and the interior of the pellet cannot be baked. Hence this is undesirable.

With the artificial lightweight aggregate of the present invention, the reason for adding a melting point lowering agent so that the total weight of the conversion amount of  $\text{Na}_2\text{O}$  and  $\text{K}_2\text{O}$  becomes 2 wt. % ~ 6 wt. % in the baked product, is because the chemical composition of the fly ash differs depending on the type of carbonaceous material, and comprises  $\text{SiO}_2$ : 50 wt. % ~ 55 wt. %,  $\text{Al}_2\text{O}_3$ : 25 wt. % ~ 30 wt. %,  $\text{Na}_2\text{O}$ : 0.2 wt. % ~ 2 wt. %,  $\text{K}_2\text{O}$ : 0.2 wt. % ~ 1 wt. %, and by adding the alkali metal group in the beforementioned amount, the melting point is greatly reduced, and the melting temperature range extended.

If the total amount of  $\text{Na}_2\text{O}$  and  $\text{K}_2\text{O}$  in the artificial lightweight aggregate falls below 2 wt. %, the baking temperature becomes 1250°C or greater, so this is not practical. Furthermore, if increased above 6 wt. %, the reduction effect on the melting point is minimal, and manufacturing costs are increased due to the increase in additives. Hence this is undesirable.

Next is a description of the foaming agent for obtaining the first artificial lightweight aggregate.

When water is added to the fly ash for pelletization, then depending on the pelletization method also, the bulk specific gravity of the dried pellet becomes 1.5 ~

1.9 approximately. When this pellet is baked at  $1000^{\circ}\text{C} \sim 1250^{\circ}\text{C}$ , the absolute dry specific gravity becomes approximately 1.5 ~ 2.0. Consequently, in order to make the absolute dry specific gravity of the artificial lightweight aggregate 0.5 ~ 1.5 approximately, a foaming agent is added to the fly ash.

For the iron oxide of the foaming agent, a hematite with a high degree of oxidation is desirable. The reason for having the particle size of the iron oxide 10 microns or less is to promote the deoxidization reaction due to the carbonaceous material and silicon carbide during baking. Furthermore, the reason for making the amount of  $\text{Fe}_2\text{O}_3$  in the artificial aggregate 1 wt. % or more during baking is because if less than this, the effect as a foaming agent is minimal, and the absolute dry specific gravity of the artificial aggregate cannot be reduced to 1.0 ~ 1.5 approximately. Furthermore, in order to make the absolute dry specific gravity 0.5 ~ 1.0, the  $\text{Fe}_2\text{O}_3$  amount must be made 3 wt. % or greater so that the silicon carbide is adequately reacted. Due to the foaming action from the carbon produced by dissociation of the silicon carbide, the lightening is considerable. On the other hand, even if the amount of  $\text{Fe}_2\text{O}_3$  in the baked aggregate exceeds 10 wt %, the lightening effect due to the foaming does not increase. Here the specific gravity of the iron oxide is very much greater than that of the fly ash, and if foaming is not promoted, the absolute dry specific gravity of the artificial lightweight aggregate is increased.

When liquid phase is produced in large amounts by heating granulated pellets, the silicon carbide reacts with iron oxide ( $\text{Fe}_2\text{O}_3$ ) with good efficiency to produce CO and  $\text{CO}_2$  gas. This CO and  $\text{CO}_2$  gas is captured and promotes the swelling of

the bubbles in the pellet. With the amount of silicon carbide less than 0.1 wt%, then the lightening effect for an absolute dry specific gravity is 0.5 ~ 1.0 is not sufficient, and an absolute dry specific gravity of 1.0 or less cannot be attained. On the other hand, even if this exceeds 10 wt%, the lightening effect is not increased.

With the carbonaceous material, the effect of adjusting the degree of reduction inside the pellet during sintering as discussed later is great, and also this reacts with the iron oxide to achieve a foaming action.

Next is a description of a material for adjusting the degree of reduction in order to obtain a second artificial lightweight aggregate.

The reason for maintaining the interior of the granulated pellet in a reducing atmosphere due to the added carbonaceous material is to reduce the hematite, being the iron oxide contained in the fly ash, to wustite or magnetite to thus lower the melting point of the matrix, to oxidize the pellet surface, and to increase the fire resistance to mitigate the fusion of the pellet at the time of heating, to increase the baking temperature, and to promote the sintering of the interior to increase the aggregate strength and reduce water absorption.

With the added proportion of the carbonaceous material of 0.2 wt % or less, conditions for reducing the interior of the pellet cannot be maintained so that the effect of lowering the melting point inside the pellet cannot be obtained. Furthermore, if the added proportion of the carbonaceous material exceeds 10 wt%, then unburned carbon will remain in the pellet interior, and since the reactivity of this residual carbon with silicates is poor, there is the possibility of a reduction in the

strength of the artificial lightweight aggregate and an increase in water absorption. Hence this is undesirable.

When water is added to the fly ash for pelletization, then depending on the pelletization method also, the bulk specific gravity of the dried pellet becomes 1.5 ~ 1.9 approximately. If the pellet is baked at 1000°C ~ 1250°C together with a material for adjusting the degree of reduction, then a high strength aggregate of an absolute dry specific gravity about 1.5 ~ 2.0 is obtained with some baking shrinkage.

### Examples

The present invention will now be described using the following working examples.

Working examples 1 ~ 65 are for the first artificial lightweight aggregate, while working examples 66 ~ 94 are for the second artificial lightweight aggregate. The chemical compositions of the fly ash, bentonite, hematite, silicon carbide, and coke used in the experiments are shown in Table 1. Furthermore, the melting point lowering agent was made by mixing the fly ash shown in Table 1 with a reagent first grade product of sodium carbonate and potassium carbonate as the alkali metal raw material, heating in an electric furnace under the conditions shown in Table 2, at a predetermined temperature for 10 minutes, removing from the furnace and cooling, and then pulverizing.

[Working Examples 1 ~ 31: foaming with hematite and carbonaceous material]

The beforementioned raw materials were collected and weighed in the composition shown in Table 3, and then pulverized and mixed in a ball mill. The particle size distribution of the pulverized raw material was measured by a laser diffraction type particle size distribution meter, and is shown in Table 3.

With the addition of water to the obtained pulverized raw material, this was pelletized to a spherical shape of approximately 5 ~ 15mm diameter in a pan pelletizer, and then dried, after which the pellets were fed to a rotary kiln (brick lining internal diameter 500mm and length 4800mm) and baked. The chemical composition of the alkali metal in the post baked artificial lightweight aggregate is shown in Table 3.

The absolute dry specific gravity and the water absorption of the baked artificial lightweight aggregate was measured based on JIS A 1110, and the crushing strength was measured for an artificial lightweight aggregate of approximately 10mm in diameter. The obtained results and baking temperatures are shown in Table 4. The absolute dry specific gravity was approximately 1.0 ~ 1.5, and hence an artificial lightweight aggregate of almost the same 1.2 ~ 1.4 absolute dry specific gravity of commercial artificial lightweight aggregate was obtained. Furthermore, the crushing strength at an absolute dry specific gravity of 1.2 ~ 1.3 was 11N ~ 15N compared to 5N ~ 6N for the commercial artificial lightweight aggregate, and even with the absolute dry specific gravity close to 1.0, was still 7N ~ 8N, giving an extremely high specific strength artificial lightweight aggregate. Twenty four hour water absorption was also shown low at approximately 5%.

[Comparative Examples 1, 7, 13]

In the case where the melting point lowering agent was minimal and the total weight of the alkali metal compound in the baked artificial lightweight aggregate was less than 2 wt. %, even if the baking temperature was increased to 1210°C ~ 1260°C, baking of the pellet was insufficient so that the absolute dry specific gravity exceeded 1.55, being higher than the target value (1.5), the crushing strength was low, and the water absorption was high.

[Comparative Examples 2, 8, 14]

In the case where the melting point lowering agent was abundant and the total weight of the alkali metal compound in the baked artificial lightweight aggregate exceeded 6 wt. %, the pellet surface melted at a low temperature and the baking temperature dropped to 1050°C ~ 1120°C so that the pellet interior could not be adequately baked. Hence the crushing strength dropped to 2N ~ 4N and the water absorption increased to 10% ~ 11%.

[Comparative Examples 3, 9, 15]

Even with the total weight of the alkali metal compound at 2 ~ 6 wt. %, in the case where the added amount of the hematite was minimal, while the strength was increased and the water absorption also reduced, the absolute dry specific gravity exceeded 1.55, being higher than the target value (1.5). Hence lightening was

inadequate.

[Comparative Examples 4, 10, 16]

With a total weight of alkali metal compound of 2 ~ 6 wt. %, even if the added weight of the hematite exceeded 10 wt. %, there was no improvement effect for the specific gravity, the strength, or water absorption.

[Comparative Examples 5, 11, 17]

In the case where carbonaceous material was not added, sintering was not promoted so that the absolute dry specific gravity was high, giving high water absorption at low strength.

[Comparative Example 6, 12, 18]

If the added amount of carbonaceous material exceed 10 wt%, the absolute dry specific gravity increased to approximately 1.65 and the specific strength dropped to 6N ~ 8N.

[Working Examples 32 ~ 65: carbonaceous material and silicon carbide synergistic lightening]

The beforementioned raw materials were collected and weighed in the composition shown in Table 5, and then pulverised and mixed in a ball mill. The particle size distribution of the pulverised raw material was measured by a laser

diffraction type particle size distribution meter, and is shown in Table 5.

With the addition of water to the obtained pulverized raw material, this was pelletized to a spherical shape of approximately 5 ~ 15mm diameter in a pan pelletizer, and then dried, after which the pellets were fed to a rotary kiln (brick lining internal diameter 500mm and length 4800mm) and baked. The included amount of the alkali metals and the included amount of iron converted  $\text{Fe}_2\text{O}_3$  in the post baked artificial lightweight aggregate is shown in Table 5.

The specific gravity and the water absorption of the baked artificial lightweight aggregate was measured based on JIS A 1110, and the crushing strength was measured for an artificial aggregate of approximately 10mm in diameter. The obtained results and baking temperatures are shown in Table 6. The absolute dry specific gravity was approximately 0.5 ~ 1.0, and hence an extremely light artificial lightweight aggregate was obtained. Furthermore, the crushing strength close to an absolute dry specific gravity of 0.5 was 3N, however at close to an absolute dry specific gravity of 1.0, this was 7N ~ 8N, giving an extremely high specific strength artificial lightweight aggregate. Twenty four hour water absorption was 12% ~ 13% at an absolute dry specific gravity of 0.5, and was 6% at an absolute dry specific gravity of 1.0.

#### [Comparative Example 19]

In the case where the melting point lowering agent was minimal and the total weight of the alkali metal compound in the baked artificial lightweight aggregate

was less than 2 wt. %, even if the baking temperature was increased to 1300°C, baking of the pellet was insufficient so that irrespective of addition of foaming agent, the absolute dry specific gravity was high at 1.21, the crushing strength was low at 2.2N, and the water absorption increased to 14.2.

[Comparative Example 20]

In the case where the melting point lowering agent was abundant and the total weight of the alkali metal compound in the baked artificial lightweight aggregate exceeded 6 wt. %, the pellet surface melted at a low temperature and the baking temperature dropped to 1000°C so that the pellet interior could not be adequately baked. Hence the absolute dry specific gravity increased to 1.37 being above the target value (1.0), the crushing strength dropped to 2.7N and the water absorption increased to 13.1%.

[Comparative Example 21]

Even with the total weight of the alkali metal compound at 2 ~ 6 wt. %, and the added amount of the hematite abundant such that the included proportion of  $\text{Fe}_2\text{O}_3$  after baking exceeded 10%, there was no significant change in the absolute dry specific gravity or the strength, and the effect of increasing the amount of hematite was not apparent.

[Comparative Example 22]

In the case where silicon carbide was not added, the absolute dry specific gravity became 1.15, not falling to the target value (1.0). Here this example belongs to the working example where deoxygenation of the hematite was carried out with carbonaceous material only.

[Comparative Example 23]

Even if the additive amount of silicon carbide exceeded 1 wt. %, the effect of lowering the absolute dry specific gravity was not improved.

[Comparative Example 24]

In the case where coke (carbonaceous material) was not added at all, oxidation inside the artificial lightweight aggregate progressed so that swelling of the bubbles was minimal and the absolute dry specific gravity became 1.44, falling short of the target value (1.0).

[Comparative Example 25]

If coke (carbonaceous material) was added at more than more than 1.0 wt. %, the degree of oxidation of the surface of the artificial lightweight aggregate became minimal so that the baking temperature could not be increased. Hence the specific gravity increased and the strength dropped.

[Working Examples 66 ~ 94]

The beforementioned raw materials were collected and weighed in the composition shown in Table 7, and then pulverized and mixed in a ball mill. The particle size distribution of the pulverized raw material was measured by a laser diffraction type particle size distribution meter, and is shown in Table 7.

With the addition of water to the obtained pulverized raw material, this was pelletized to a spherical shape of approximately 5 ~ 15mm diameter in a pan pelletizer, and then dried, after which the pellets were fed to a rotary kiln (brick lining internal diameter 500mm and length 4800mm) and baked. The chemical composition of the alkali metal in the post baked artificial lightweight aggregate is shown in Table 7. The specific gravity and the water absorption of the baked artificial lightweight aggregate was measured based on JIS A 1110, and the crushing strength was measured for an artificial lightweight aggregate of approximately 10mm in diameter. The obtained results and baking temperatures are shown in Table 8. The absolute dry specific gravity was approximately 1.5 ~ 2.0. Furthermore, the crushing strength was 15N ~ 40N compared to 5N ~ 6N for the commercial artificial lightweight aggregate, giving an extremely high specific strength artificial lightweight aggregate. Twenty four hour water absorption was also shown low at approximately 0.3% ~4%.

[Comparative Examples 26, 30, 34]

In the case where the melting point lowering agent was minimal and the total weight of the alkali metal compound in the baked artificial lightweight aggregate

was less than 2 wt. %, even if the baking temperature was increased to 1250°C ~ 1270°C, baking of the pellet was insufficient so that the crushing strength was low and the water absorption high.

[Comparative Examples 27, 31, 35]

In the case where the melting point lowering agent was abundant and the total weight of the alkali metal compound in the baked artificial lightweight aggregate exceeded 6 wt. %, the pellet surface melted at a low temperature so that the baking temperature could not be increased to approach 1000°C. Hence since the pellet interior could not be adequately baked, the crushing strength dropped to 5N ~ 9N and the water absorption increased to 5% ~ 8%.

[Comparative Examples 28, 32, 36]

Even with the total weight of the alkali metal compound at 2 ~ 6 wt. %, in the case where carbonaceous material (coke) was not added, the interior of the pellet was not in a reducing state and hence the formation of a liquid phase was not promoted, so that adequate strength was not obtained.

[Comparative Examples 29, 33, 37]

With a total weight of alkali metal compound of 2 ~ 6 wt. %, if the added weight of the hematite was more than 10 wt. %, there was no improvement effect for the absolute dry specific gravity, the strength, or water absorption, being a little

worse.

With the present invention, since this is constructed as described above, a high quality artificial lightweight aggregate of low cost can be efficiently produced using fly ash produced from coal fired boilers as the raw material. Consequently, the fly ash can be recycled for building materials or the like where light weight is required, instead of disposal as industrial waste in reclamations. Hence the contribution to environmental maintenance and a stable supply of energy is significant.

[Table 1]

Component	Fly Ash	Bentonite	Hematite	Silicon Carbide	Coke
SiO <sub>2</sub>	56.2	65.8	1.03	143.25	7.56
Al <sub>2</sub> O <sub>3</sub>	32.1	13.2	97.8		3.24
Fe <sub>2</sub> O <sub>3</sub>	3.57	1.55			
CaO	0.59	0.55			
MgO	1.4	1.8			
Na <sub>2</sub> O	0.22	1.59			
K <sub>2</sub> O	0.48	1.7			
SO <sub>2</sub>		0.48			0.61
C				29.06	88.3
LL		13.42			
Total	94.56	100.09	98.83	172.31	99.71

[Table 2]

Melting Point Lowering Agent No.	Fly Ash (W.%)	Sodium Carbonate (W.%)	Potassium Carbonate (W.%)	Alkali Metal after Heat treatment (W.%)	Temperature of Heat treatment (°C)
1-1	70	30	0	21.0	1200
1-2	58	42	0	31.9	1200
1-3	50	50	0	38.1	1100
1-4	40	60	0	48.0	1000
1-5	30	70	0	58.9	1000
2-1	70	0	30	23.8	1200
2-2	60	0	40	32.6	1200
2-3	50	0	50	41.9	1100
2-4	45	0	55	46.9	1000
2-5	30	0	70	62.7	1000
3-1	70	15	15	22.7	1200
3-2	60	20	20	31.1	1200
3-3	50	25	25	40.2	1100
3-4	44	28	28	46.1	1000
3-5	30	35	35	61.0	1000

[Table 3]

	Material Composition						Pellet	Particle Size
	Fly Ash	Melting Point	Bentonite	Hematite	Coke	Na <sub>2</sub> O+K <sub>2</sub> O	(μ m)	
	(W.%)	Lowing Agent	(W.%)	(W.%)	(W.%)	(W.%)		
	No.	Ratio						
Example1	89		4	1	3	3	2.1	9
2	83	1-2	10	1	3	3	4	13
3	78		15	1	3	3	5.6	9
Comparison								
Example1	90		3	1	3	3	1.7	13
2	76		17	1	3	3	6.3	13
3	86	1-2	10	1	0	3	4.1	10
4	74		10	1	12	3	3.9	14
5	86		10	1	3	0	4.1	9
6	75		10	1	3	11	4.3	11
Example4	89		4	1	3	3	2.4	9
5	85		8	1	3	3	4	9
6	81		12	1	3	3	5.7	12
7	87	1-3	8	1	1	3	4	13
8	78		8	1	10	3	3.9	11
9	87		8	1	3	1	3.9	8
10	78		8	1	3	10	4.2	7
C-E	7	91	2	1	3	3	1.6	11
	8	79	14	1	3	3	6.4	12
	9	88	8	1	0	3	4	15
	10	76	8	1	12	3	3.9	10
	11	88	8	1	3	0	3.9	12
	12	77	8	1	3	11	4.2	12
Example11	90		3	1	3	3	2.3	14
12	87	1-4	6	1	3	3	3.8	10
13	83		10	1	3	3	5.8	10
C-E	13	91	2	1	3	3	1.8	12
	14	88	12	1	3	3	6.9	10
	15	90	6	1	0	3	3.9	12
	16	78	6	1	12	3	3.7	11
	17	90	6	1	3	0	3.7	7
	18	79	6	1	3	11	4	10
Example14	89		4	1	3	3	2.1	9
15	84	2-2	9	1	3	3	3.9	10
16	78		15	1	3	3	5.9	10

[Table 3] continued.

Example17	90		3	1	3	3	2.1	12
18	86	2-3	7	1	3	3	3.9	11
19	82		11	1	3	3	5.6	12
Example20	90		3	1	3	3	2.2	12
21	86	2-4	7	1	3	3	4.2	11
22	83		10	1	3	3	5.7	9
Example23	89		4	1	3	3	2.1	10
24	83	3-2	10	1	3	3	4.	8
25	78		15	1	3	3	5.7	11
Example26	90		3	1	3	3	2	11
27	85	3-3	8	1	3	3	4.2	11
28	81		12	1	3	3	3.9	10
Example29	90		3	1	3	3	2.2	11
30	86	3-4	7	1	3	3	4.2	13
31	83		10	1	3	3	5.7	10

\*C-E: Comparison Example

[Table 4]

	Absolute Dry Specific Gravity	Crushing Strength (N)	Water Absorption (24H) (D.B.%)	Baking Temperature (°C)
Example1	1.44	18	4.7	1230
2	1.13	10	5.8	1180
3	0.98	7	6.1	1150
Comparison				
Example1	1.58	3	8.8	1260
2	1.61	3	10.2	1120
3	1.78	21	4.3	1200
4	1.22	12	5.4	1160
5	1.55	4	8.1	1200
6	1.63	8	5.7	1110
Example4	1.39	16	4.8	1200
5	1.15	10	5.7	1160
6	1.10	9	6.0	1120
7	1.26	13	5.2	1190
8	1.11	10	5.8	1140
9	1.26	12	5.3	1170
10	1.29	15	5.0	1150
C- Example7	1.62	3	9.7	1240
8	1.66	2	11.4	1070
9	1.74	23	4.3	1180
10	1.19	11	5.5	1140
11	1.57	4	8.4	1180
12	1.65	6	6.7	1140
Example11	1.51	21	4.5	1180
12	1.33	15	5.0	1140
13	1.26	11	5.4	1110
C-Example13	1.60	2	11.1	1210
14	1.81	4	7.9	1050
15	1.59	19	4.5	1160
16	1.27	11	5.4	1120
17	1.58	3	9.5	1150
18	1.64	6	6.4	1130
Example14	1.42	18	4.7	1190
15	1.12	9	5.9	1150
16	1.05	8	6.2	1120
17	1.39	17	4.7	1170
18	1.12	10	5.8	1130

[Table 4] continued.

19	0.95	9	6.0	1100
20	1.50	20	4.5	1150
21	1.23	12	5.3	1110
22	1.19	11	5.5	1070
23	1.43	19	4.6	1200
24	1.14	9	5.9	1160
25	1.06	8	6.3	1130
26	1.45	20	4.6	1180
27	1.14	10	5.8	1140
28	1.12	9	5.9	1100
29	1.53	23	4.4	1160
30	1.26	13	5.2	1100
31	1.21	11	5.4	1080

[Table 5]

TABLE 57

	Material Composition					Pellet		Particle		
	Fly Ash	Melting Point	Bentonite	Hematite	Silicon	Coke	Na <sub>2</sub> O+K <sub>2</sub> O	Fe <sub>2</sub> O <sub>3</sub>	Size	
	(W.%)	Lowing Agent	(W.%)	(W.%)	Carbide	(W.%)	(W.%)	(W.%)	( $\mu$ m)	
		No.	Ratio		(W.%)					
E-32	86.5		4	1	3	0.5	5.0	2.1	6.8	12
33	80.5	1-2	10	1	3	0.5	5.0	4.1	6.7	12
34	75.5		15	1	3	0.5	5.0	5.8	6.6	14
E-35	86.5		4	1	3	0.5	5.0	2.4	6.8	12
36	77.9		10	1	1	0.1	10.0	5.1	4.6	12
37	82.5		10	1	1	0.5	5.0	4.9	4.6	11
38	86.8		10	1	1	1.0	0.2	4.7	4.6	10
39	75.9	1-3	10	1	3	0.1	10.0	5.1	6.8	14
40	80.6		10	1	3	0.5	5.0	4.9	6.7	14
41	84.8		10	1	3	1.0	0.2	4.7	6.6	11
42	72.9		10	1	6	0.1	10.0	5.0	10.0	12
43	77.5		10	1	6	0.5	5.0	4.9	9.8	13
44	81.8		10	1	9	1.0	0.2	4.7	9.5	12
45	76.5		15	1	3	0.5	5.0	6.9	6.6	10
C-19	88.5		2	1	3	0.5	5.0	1.6	6.8	14
20	72.5		18	1	3	0.5	5.0	8.2	6.5	11
21	86.5		10	1	7	0.5	5.0	4.8	10.8	11
22	81.0	1-3	10	1	3	0.0	5.0	4.9	6.7	15
23	79.8		10	1	3	1.2	5.0	4.9	6.7	10
24	85.5		10	1	3	0.5	0.0	4.7	6.6	12
25	73.5		10	1	3	0.5	12.0	5.2	6.8	12
E-46	86.5		4	1	3	0.5	5.0	2.8	6.8	11
47	82.5	1-4	8	1	3	0.5	5.0	4.9	6.7	14
48	78.5		12	1	3	0.5	5.0	7.0	6.6	13
E-49	86.5		4	1	3	0.5	5.0	2.1	6.8	13
50	80.5	2-2	8	1	3	0.5	5.0	4.3	6.7	13
51	75.5		15	1	3	0.5	5.0	6.0	6.6	11
E-52	86.5		4	1	3	0.5	5.0	2.6	6.8	11
53	77.9		10	1	1	0.1	10.0	5.5	4.6	14
54	82.5		10	1	1	0.5	5.0	4.9	4.6	13
55	86.8		10	1	1	1.0	0.2	5.1	4.6	12
56	75.9		10	1	3	0.1	10.0	5.5	6.8	10
57	80.5	2-3	10	1	3	0.5	5.0	5.3	6.7	13
58	84.8		10	1	3	1.0	0.2	5.1	6.5	12
59	72.9		10	1	6	0.1	10.0	5.5	10.0	15

[Table 5] continued.

60	77.5		10	1	6	0.5	5.0	5.3	9.8	13
61	81.8		10	1	6	1.0	0.2	5.1	9.5	14
62	75.5		15	1	3	0.5	5.0	7.5	6.6	10
E-63	86.5		4	1	3	0.5	5.0	2.8	6.8	12
64	82.5	2-4	8	1	3	0.5	5.0	4.8	6.7	12
65	78.5		12	1	3	0.5	5.0	6.8	6.6	12

\* E: Example

C: Comparison Example

[Table 6]

	Absolute Dry Specific Gravity	Crushing Strength (N)	Water Absorption (24H) (D.B.%)	Baking Temperature (°C)
Example32	0.69	4.8	9.2	1250
33	0.51	3.0	11.8	1050
34	0.56	3.5	10.9	1020
Example35	0.72	5.1	8.8	1190
36	0.97	7.6	6.3	1070
37	0.88	6.4	7.2	1070
38	0.75	5.2	9.0	1080
39	0.89	6.2	8.1	1030
40	0.53	3.2	11.5	1030
41	0.52	3.0	12.7	1030
42	0.91	7.0	8.9	1020
43	0.48	2.9	12.3	1020
44	0.50	2.8	13.0	1030
45	0.54	3.3	11.4	1020
C-E 19	1.21	2.2	14.2	1300
20	1.37	9.7	3.1	1000
21	0.56	3.5	11.1	1030
22	1.15	10.1	4.7	1030
23	0.53	3.2	11.5	1030
24	1.44	5.0	16.7	1030
25	0.67	3.4	11.6	1030
Example46	0.85	6.6	7.3	1130
47	0.63	4.3	10.1	1030
48	0.78	5.3	8.5	1020
Example49	0.64	4.2	9.0	1250
50	0.50	3.2	11.0	1040
51	0.57	3.6	12.0	1020
Example52	0.72	5.3	8.6	1160
53	0.97	6.6	6.7	1070
54	0.88	6.6	7.2	1070
55	0.75	4.2	9.0	1070
56	0.89	5.7	8.1	1020
57	0.53	3.2	11.5	1030
58	0.52	2.8	12.7	1030
59	0.91	5.9	6.9	1020
60	0.48	2.9	12.1	1020

[Table 6] continued.

61	0.50	2.6	13.0	1020
62	0.54	3.3	11.4	1020
Example 63	0.79	6.0	8.4	1130
64	0.62	4.0	10.1	1030
65	0.72	4.7	8.6	1020

\* C-E: Comparison Example

[Table 7]

		Material Composition			Pellet		Particle Size ( $\mu$ m)
		Fly Ash	Melting Point	Bentonite	Coke	Na <sub>2</sub> O+K <sub>2</sub> O	
		(W.%)	Lowing Agent No. Ratio	(W.%)	(W.%)	(W.%)	
Example66	92		4	1	3	2.1	11
	67	86	1-2	10	1	3	4.1
	68	81		15	1	3	5.7
C-E	26	93		3	1	3	1.8
	27	79		17	1	3	6.2
	28	89	1-2	10	1	0	4
	29	78		10	1	11	4.3
Example69	92		4	1	3	2.4	9
	70	88		8	1	3	4
	71	84		12	1	3	5.7
	72	90	1-3	8	1	1	4
	73	81		8	1	10	4.2
C-E	30	94		2	1	3	1.6
	31	82		14	1	3	6.5
	32	91		8	1	0	4
	33	80	1-3	10	1	11	4.3
Example74	93		3	1	3	2.3	8
	75	90	1-4	6	1	3	3.9
	76	86		10	1	3	5.9
C-E	34	94		2	1	3	1.8
	35	84		12	1	3	6.9
	36	93	1-4	6	1	0	3.8
	37	82		6	1	11	4.1
Example77	92		4	1	3	2.2	7
	78	87	2-2	9	1	3	3.9
	79	81		15	1	3	5.9
Example80	93		3	1	3	2.2	11
	81	89	2-3	7	1	3	3.9
	82	85		11	1	3	5.6
Example83	90		3	1	3	2.2	8
	84	89	2-4	7	1	3	4.3
	85	86		10	1	3	5.8
Example86	92		4	1	3	2.1	10
	87	86	3-2	10	1	3	4
	88	81		15	1	3	5.7

[Table 7] continued.

Example89	93		3	1	3	2	10
90	88	3-3	8	1	3	4.2	11
91	84		12	1	3	5.9	14
Example92	93		3	1	3	2.3	11
93	89	3-4	7	1	3	4.2	9
94	86		10	1	3	5.7	7

\* C-E: Comparison Example

[Table 8]

		Absolute Dry Specific Gravity	Crushing Strength (N)	Water Absorption (24H) (D.B.%)	Baking Temperature (°C)
Example 66		1.89	37	0.9	1240
	67	1.78	28	1.9	1130
	68	1.52	15	3.9	1000
C-E	26	1.63	10	6.0	1270
	27	1.57	7	7.1	1010
	28	1.80	4	3.6	1140
	29	1.74	19	2.6	1140
Example 69		1.97	36	0.3	1230
	70	1.80	28	1.8	1150
	71	1.61	16	3.0	1050
	72	1.79	29	1.7	1140
	73	1.73	27	2.1	1130
C-E	30	1.84	8	4.5	1270
	31	1.57	5	5.5	1000
	32	1.70	4	5.0	1150
	33	1.65	13	4.2	1110
Example 74		1.94	37	0.5	1220
	75	1.75	28	2.0	1130
	76	1.56	18	3.6	1030
C-E	34	1.79	10	3.9	1250
	35	1.58	9	7.8	990
	36	1.78	3	4.0	1140
	37	1.65	25	3.7	1130
Example 77		1.93	37	0.6	1230
	78	1.76	28	2.3	1130
	79	1.54	17	3.5	1020
	80	1.92	36	0.7	1230
	81	1.77	27	2.0	1130
	82	1.59	19	3.3	1050
	83	1.95	38	0.5	1230
	84	1.78	31	1.7	1130
	85	1.71	24	2.4	1130
	86	1.97	39	0.3	1240
	87	1.78	26	2.2	1140
	88	1.60	22	3.1	1050
	89	2.00	40	0.2	1250

(Table 8) continued.

90	1.73	27	2.5	1130
91	1.59	20	3.5	1030
92	1.76	27	1.7	1130
93	1.73	25	2.3	1120
94	1.61	19	3.2	1060

\* C-E: Comparison Example

## Claims

1. An artificial lightweight aggregate made by mixing fly ash with a melting point lowering agent, a caking agent, and a foaming agent to obtain a mixture, crushing said mixture so that the average particle size is up to 15 microns to obtain a pulverized product, forming small bodies of said pulverized product, and then baking said small bodies within a temperature range of 1000°C ~ 1250°C to produce the aggregate with an absolute dry specific gravity of 1.0 ~ 0.5.
2. A method of manufacturing artificial lightweight aggregate comprising the steps of; mixing fly ash with a melting point lowering agent, a caking agent, and carbonaceous material to obtain a mixture, crushing said mixture to obtain a pulverized product with the average particle size up to 15 microns, forming small bodies of said pulverized product, and then baking said small bodies within a temperature range of 1000°C ~ 1250°C.
3. A method of manufacturing artificial lightweight aggregate according to claim 2, wherein said carbonaceous material is within a range of 0.2 wt. % ~ 10 wt. % of the fly ash.
4. A method of manufacturing artificial lightweight aggregate according to claim 2, wherein in addition, iron oxide is mixed in the mixture.
5. A method of manufacturing artificial lightweight aggregate according to claim 4, wherein in addition hydrogen carbonate is mixed in the mixture.
6. A method of manufacturing artificial lightweight aggregate according to either one of claim 4 and claim 5, involving iron oxide in an amount such that the amount

of  $\text{Fe}_2\text{O}_3$  in the fly ash is within the range of 1 wt. % ~ 10 wt. %, carbonaceous material in an amount within the range of 0.2 wt. % ~ 10 wt. % of the fly ash, and silicon carbide in an amount within the range of 0 wt. % ~ 1 wt. % of the fly ash.

7. A method of manufacturing artificial lightweight aggregate according to any one of claim 2 through claim 6, wherein the melting point lowering agent is made by mixing an alkali metal compound with fly ash so that the total amount of  $\text{Na}_2\text{O}$  and  $\text{K}_2\text{O}$  respectively or both is within the range of 30 wt. % ~ 50 wt. % in the mixture, heating and melting the mixture within a temperature range of  $1000^\circ\text{C}$  ~  $1200^\circ\text{C}$  to form a glassy material, and then cooling and crushing the glassy material.
8. A method of manufacturing artificial lightweight aggregate according to any one of claim 2 through claim 6, wherein said melting point lowering agent is made by mixing an alkali metal compound with fly ash so that the total amount of  $\text{Na}_2\text{O}$  and  $\text{K}_2\text{O}$  respectively or both is within a range of 30 wt. % ~ 50 wt. % in the mixture, heating and melting the mixture within a temperature range of  $1000^\circ\text{C}$  ~  $1200^\circ\text{C}$  to form a glassy material, and then cooling and crushing the glassy material, and said melting point lowering agent is added to the fly ash so that the total amount of  $\text{Na}_2\text{O}$  and  $\text{K}_2\text{O}$  is within the range of 2 wt. % ~ 6 wt. % of the baked product.
9. A method of manufacturing artificial lightweight aggregate according to either one of claim 7 and claim 8, wherein said alkali metal compound is sodium carbonate and potassium carbonate.
10. A method of manufacturing artificial lightweight aggregate according to claim 2, wherein said small bodies are pellets, and a rotary kiln is used as the baking

furnace.

11. A melting point lowering agent for an artificial lightweight aggregate made by mixing an alkali metal compound with fly ash so that the total amount of  $\text{Na}_2\text{O}$  and  $\text{K}_2\text{O}$  respectively or both is within a range of 30 wt. % ~ 50 wt. % in the mixture, heating and melting the mixture within a temperature range of  $1000^\circ\text{C}$  ~  $1200^\circ\text{C}$  to form a glassy material, and then cooling and crushing the glassy material.
12. A melting point lowering agent for an artificial lightweight aggregate according to claim 11, wherein said alkali metal compound is sodium carbonate and potassium carbonate.
13. An artificial lightweight aggregate as hereinbefore described.
14. A method of manufacturing an artificial lightweight aggregate as hereinbefore described.



Application No: GB 9821749.0  
Claims searched: 1 to 10,13 and 14

Examiner: Miss M M Kelman  
Date of search: 18 January 1999

**Patents Act 1977**  
**Search Report under Section 17**

**Databases searched:**

UK Patent Office collections, including GB, EP, WO & US patent specifications, in:  
UK CI (Ed.Q): C1A APF5 APF8 APHS, C1H HAV HAW  
Int CI (Ed.6): C04B 18/00, 18/04, 18/06, 18/08, 18/10, 20/00, 20/02, 20/04  
Other: ONLINE: EDOC, WPI

**Documents considered to be relevant:**

Category	Identity of document and relevant passage	Relevant to claims
A	GB 1417685 A CHARMONNAGES DE FRANCE	1,2,3,4,10 ,13,14 at least
A	WPI Abstract Accession No. 93-352214[45] & AU 003528293 A (LIGHTWEIGHT BLOCKS) 23 September 1993 see abstract	
X,P	WPI Abstract Accession No. 98-280314[25] & JP 100095648 A (SUMITOMO) 14 April 1998 see abstract	
A	WPI Abstract Accession No. 96-483742[48] & KR 950001668 B1 (LEE) 28 February 1995 see abstract	

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